Journal of Environmental Management 222 (2018) 164–173

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Photocatalytic-oxidation and photo-persulfate-oxidation of sulfadiazine in a laboratory-scale reactor: Analysis of catalyst support, oxidant dosage, removal-rate and degradation pathway

M S Priyanka Yadav[a], N. Neghi[b], Mathava Kumar[b,*], George K. Varghese[a]

[a] Department of Civil Engineering, National Institute of Technology, Calicut, Kerala, 673601, India
[b] Department of Civil Engineering, Indian Institute of Technology Madras, Chennai, Tamilnadu, 600036, India

ARTICLE INFO

Keywords:
Advanced oxidation
Persulfate
Photocatalysis
Sulfadiazine
Wastewater treatment

ABSTRACT

The extent of sulfadiazine (SDZ) removal via photo-degradation (UV-C), photocatalysis with TiO_2 (UV-C/TiO_2) and photo-persulfate-oxidation (UV-C/PS) was investigated in a batch reactor under different UV-C power levels (i.e. 14, 28, 42 and 56 W). Moreover, effects of suspended/immobilized catalyst, i.e. TiO_2 slurry/TiO_2 supported on granular activated carbon (GAC-TiO_2), on SDZ removal and corresponding SDZ degradation kinetics under different catalyst loading (1–6 g/L) were explored. Around 41.7% SDZ removal was observed after 120 min in UV-C system at the highest power level, i.e. 56 W. On the other hand, photocatalysis with TiO_2 and GAC-TiO_2 has shown better SDZ removal than photo-degradation. In UV-C/TiO_2 (4 g/L and 28 W) and UV-C/GAC-TiO_2 (5 g/L and 28 W) systems, SDZ removals were 91.8% after 120 min and 100% after 60 min, respectively; however, TOC analysis has revealed that 45.4% and 60.8% SDZ was mineralized in these systems, respectively. In UV-C/PS system, near complete degradation of SDZ (99.8%) was observed within 10 min under 50 mg/L of PS and 28 W UV illumination. On the other hand, complete SDZ removal was observed in PS alone system at a dosage of 1000 mg/L but the formation of SO_4^{2-} was found to be a drawback. In photolysis and photocatalysis systems, SDZ removal followed pseudo-first-order kinetics whereas the kinetics followed pseudo-second-order in UV-C/PS system. The comparison of electrical energy consumed (E_{EO}) in different systems revealed that UV-C/GAC-TiO_2 and UV-C/PS system were energy efficient compared with other systems. The LC-MS analysis has confirmed the cleavage of C-N bonds in the pyrimidine ring followed by S-N bonds in the sulfonyl group, which was found to be the major degradation pathway of SDZ.

1. Introduction

Sulfadiazine (SDZ, $C_{10}H_{10}N_4O_2S$) belongs to the group of sulfonamides and it is one of the most commonly used human and veterinary antibiotics. SDZ has moderate solubility in water (77 mg/L), long persistence and high mobility (Gros et al., 2013; Sukul et al., 2008). SDZ is considered as one of the potential compounds that can produce threat to human life and environment (Rong et al., 2014). A traceable level of SDZ was observed in various aqueous environments including soil (4130 ng/L), surface water (1000 ng/L) and industrial effluents (353 ng/L) (Akhtar et al., 2016; Kemper, 2008). In addition to the above environmental compartments, SDZ was also found in pig manure (63.4 mg/L). The application of pig manure for agricultural purposes could potentially lead to several environmental issues (Sukul et al., 2008). It was reported that one of the significant metabolite of SDZ, i.e. N-acetyl-sulfadiazine, converts back to parental form by hydrolysis and also it has very long half-life in the environment. Moreover, SDZ is removed partially or incompletely in the biological wastewater treatment units. It was reported earlier that around 60% of the toxic pharmaceuticals are removed in the biological treatment processes (Baran et al., 2009). As a result, the effluent from secondary settling tank tends to contain significant levels of SDZ and other pharmaceuticals (Pan et al., 2014). Although a variety of physico-chemical and biological techniques are useful in the removal of these recalcitrant compounds. Most physico-chemical methods including filtration, coagulation-flocculation, clarification, sedimentation and few adsorption techniques transfer the contaminants from one phase to another wherein complete degradation or mineralization has not been achieved (Liu et al., 2014b; Manjunath et al., 2017). On the other hand, biological methods like activated sludge process can be employed to the effluents with low toxicity and low concentrations of recalcitrant compounds (Homem and Santos, 2011). The limitations of physical and biological processes to

* Corresponding author.
E-mail address: mathav@iitm.ac.in (M. Kumar).

https://doi.org/10.1016/j.jenvman.2018.05.052
Received 2 January 2018; Received in revised form 8 May 2018; Accepted 16 May 2018
0301-4797/ © 2018 Elsevier Ltd. All rights reserved.

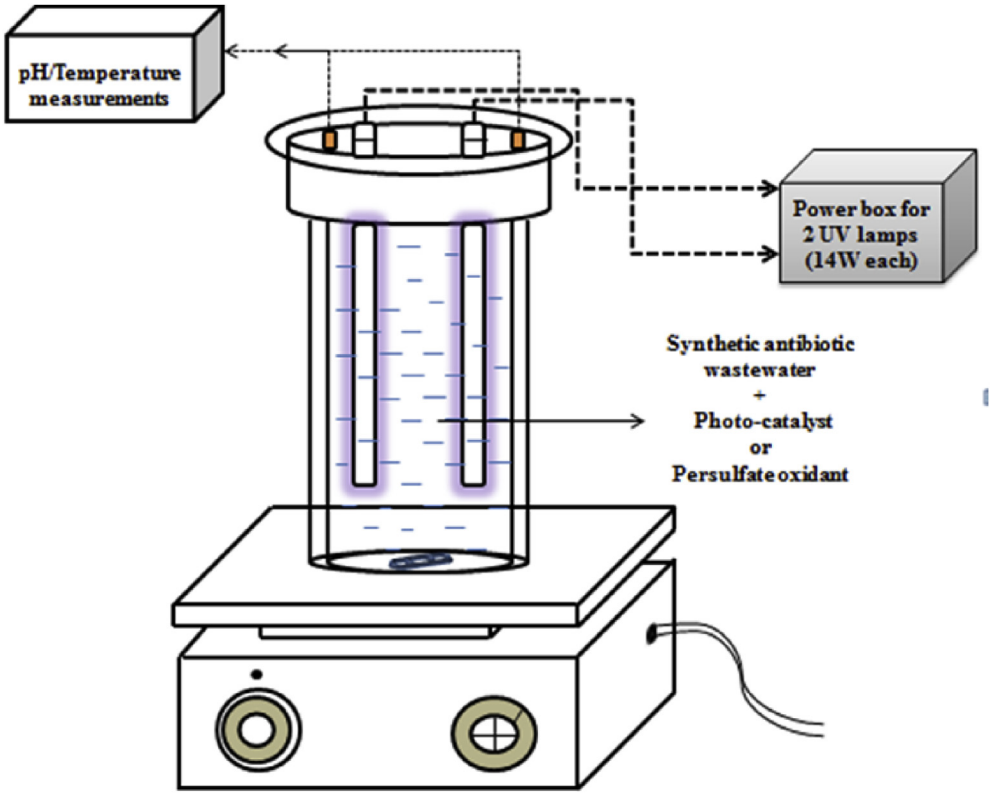

Fig. 1. Schematic of batch photo-catalytic reactor.

defend or act as barrier for various organic pollutants including pharmaceutical compounds paved way for the application of advanced oxidation processes (AOP's).

AOP's are considered to be one of the cleaner and promising technologies to treat the wastewater containing oxidizable and recalcitrant organic pollutants. The removal of pollutants is achieved by the generation of highly reactive and non-selective $OH^{\cdot}$ radicals (Verma et al., 2014). To date, wide variety of AOP's were developed including photolysis, photocatalysis, Fenton process, ozonation, radiolysis and electrochemical oxidation (Liu et al., 2014b). AOP's utilize different combinations of irradiation (solar light and ultraviolet light), oxidants such as ozone, hydrogen peroxide (H_2O_2), transition metal and metal oxides, electric current, gamma-radiation, ultrasound and catalysts for the generation of $OH^{\cdot}$ radicals in the system (Ikehata et al., 2006). Few combinations include O_3/H_2O_2, O_3/UV, O_3/H_2O_2/UV, H_2O_2/UV, Fenton (Fe^{2+}/H_2O_2), photo- and electro-Fenton, chelating agent assisted Fenton/photo-Fenton, heterogeneous photo-oxidation using titanium dioxide and sonolysis. These technologies were operated in mere ambient atmospheric conditions and were proved to be effective in the destruction of several antibiotics. Photocatalysis under UV and visible irradiation aided with several semiconductor materials, i.e. TiO_2, ZnO, ZrO_2, CdS, MoS_2, Fe_2O_3, CdS, SnO_2, ZnS, and WO_3, have been examined for the degradation of various organic pollutants (Jodat and Jodat, 2014; Kumar and Rao, 2017; Pattanaik and Sahoo, 2014). Moreover, TiO_2 has been doped with transition metals (Fe and Co) and non-metals, and also TiO_2 surface has been modified with Nafion and phosphate for visible-light photocatalysis applications (Khaki et al., 2017; Srikanth et al., 2017; Zheng et al., 2017). Among the photocatalysts, TiO_2 has been used widely in the photocatalytic oxidation of organic pollutants and photocatalytic CO_2 reduction (Low et al., 2017; Vaiano et al., 2015). Additionally, several investigations focused on coating TiO_2 on supports such as silica, glass beads, polymers (Singh et al., 2013; Yang et al., 2006) and granular activated carbon (GAC) (Gao et al., 2011; Khalid et al., 2017) for its ease of recovery after treatment. GAC has been used as a supporting material for TiO_2, which also provides great surface area for adsorption. However, estimation of GAC-TiO_2 performance for water/wastewater treatment application is still in lime-light.

Besides the photocatalyst, there are strong oxidizing agents like H_2O_2, O_3, and persulfate (PS) for increasing the reaction rate of photo-degradation and also for complete oxidation of pollutants by generation of further $OH^{\cdot}$ radicals. PS usage in the oxidation process is a unique step in AOP's where the sulfate radicals $SO_4^{-\cdot}$ destroy the oxidizable organic pollutants in wastewater with an oxidation potential of 2.5–3.1 eV. PS can be activated by the supply of heat, transition metals, H_2O_2 and UV light. These $SO_4^{-\cdot}$ are selective and therefore, high degradation rate of target pollutants can be attained in a short reaction time (Deng et al., 2013; Wang et al., 2016). It was also reported that PS activation by UV-C (254 nm) has higher efficiency in degrading few antibiotics than H_2O_2 activation (Ye et al., 2016). The major reaction of PS in the presence of UV is shown in Eqs. (1)–(5).

$$S_2O_8^{2-} + h\nu \rightarrow 2SO_4^{2-} \quad (1)$$

$$SO_4^{-\cdot} + OH^- \rightarrow HO^{\cdot} + SO_4^{-\cdot} \quad (2)$$

$$HO^{\cdot} + HO^{\cdot} \rightarrow H_2O_2 \quad (3)$$

$$HO^{\cdot} + SO_4^{-\cdot} \rightarrow HSO_5^- \quad (4)$$

$$HO^{\cdot} + HSO_5^- \rightarrow SO_5^{-\cdot} + H_2O \quad (5)$$

In the past, very limited studies have investigated the extent of antibiotics removal by photocatalysis and photo-degradation with oxidants like PS. However, comparative evaluation of the above mentioned processes, role of catalyst dosage and effect of sorption in antibiotics removal were not investigated in detail. Therefore, the main objective of this investigation was to explore the removal of SDZ from aqueous solutions by UV-C system, UV-C photocatalysis and UV-C/PS system and compare the performance of the various systems. Additionally, the study was focused to evaluate (a) the effects of photocatalyst dosage,

Download English Version:

https://daneshyari.com/en/article/7476072

Download Persian Version:

https://daneshyari.com/article/7476072

Daneshyari.com